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Subpicoliter droplet generation based on a nozzle-free acoustic transducer

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This letter reports picoliter liquid droplet generation using an orifice-free acoustic ejector operating at its harmonic frequencies. For an acoustic ejector working at the thickness-mode resonance, the droplet size is primarily determined by the acoustic wavelength, which is proportional to the piezoelectric substrate thickness. In our design, we do not need to lap the bulk piezoelectric lead zirconate titanate (PZT) substrate or deposit high temperature processing PZT thin film, but we use harmonic frequencies of the bulk form to reduce the wavelength. The fabricated acoustic ejector with a size of 1200×1200 μm^2 has been shown to be very effective up to the ninth harmonic (180 MHz), continuously ejecting ~10 μm diameter droplets, corresponding to droplet volumes as small as 0.5 pl. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2958342]

In many biomedical applications, droplet generators are essential for liquid delivery. For example, Agilent and Rosetta use their inkjet printing technology to produce deoxyribonucleic acid probe sequences at their factories.[1] The inkjet printheads eject liquid droplets through the nozzles, and the smallest droplet size depends on the size of the nozzle.[2] Small nozzles are difficult to construct with good uniformity. Picoliter droplet generation has been demonstrated using nozzles.[3] However, in many biomedical applications, the reagents block the nozzle easily, significantly downgrading the reliability and increasing the maintenance cost. The biological/chemical precipitates can accumulate on the printhead and clog the nozzles, particularly when volatile solvents are used. Hence, nozzleless ejection is desired when the droplet size needs to be minimized without clogging problems at all. Another major drawback of the nozzle-based ejectors is the generation of undesirable satellite droplets. Since the hydrostatic pressure is used to form the droplets at the nozzle, satellite droplets are typically created along with the main droplet, affecting the printing performance.[4]

Bulk acoustic waves, when focused, can produce significantly enhanced acoustic streaming near the focal point due to the intensity magnification and are effective in generating fluid motions.[5,6] Acoustic waves can be focused through a lens[7] or a self-focused acoustic transducer[8] (SFAT) having a set of annular rings for the electrodes sandwiching a piezoelectric substrate. The focused acoustic waves have been shown to be able to eject liquid droplets without any nozzles.[9] However, the size of the droplet is limited to around 0.3 nl.[10] The ejected droplet size, primarily determined by the diameter of the focused acoustic beam, is proportional to the piezoelectric substrate thickness (or inversely proportional to the frequency). Since the ejection of smaller droplets is the key for both better printing resolution and reduced consumption of dispensed reagents, higher-frequency operation is desirable for droplet ejections. Additionally, the size of the lens (or the electrodes for SFAT) is directly related to the working frequency, and thus higher-frequency operation is needed when the transducer size needs to be reduced.

Lead zirconate titanate (PZT) has been used as the piezoelectric material for acoustic transducer due to its large electromechanical coefficient and capability to produce large acoustic power. For high-frequency thickness-mode operation (over 20 MHz), the PZT transducer needs to have a thickness less than 100 μm. Lapping of the bulk ceramic is a solution to reduce the thickness.[11] However, this induces risk of fracture as well as difficulty in handling. To avoid these issues, using thin or thick films is an alternative solution, and several fabrication processes have been developed for high-frequency transducer applications. However, while sputtering[12,13] and chemical vapor deposition[14] of PZT films require very tight process control for repeatable quality, sol-gel PZT films typically have large residual stress.[15–17] In addition to these processing difficulties, the electromechanical performance obtained with thin PZT film is generally lower than those with the same compositions in the bulk form.[18]

In this letter, we describe an effective scheme to operate acoustic transducers at higher frequencies and generate smaller droplets using a bulk PZT substrate. For focusing acoustic waves, the lens is patterned into Fresnel half-wave bands so that the transmitted acoustic waves arrive at the liquid surface in phase, constructively interfering with each other and intensifying the acoustic pressure. The focused acoustic waves can eject liquid droplets without any nozzles as shown in Fig. 1(a). For an efficient acoustic-wave generation, the piezoelectric film thickness is typically chosen to be one-half of the acoustic wavelength in the piezoelectric film for an air-backed transducer working at the thickness-mode resonance. For a PZT sheet with a thickness of 127 μm, the fundamental frequency is measured to be around 20 MHz. At this frequency, the wavelength in water is around 80 μm, which limits the smallest droplet producible to be about 80 μm in diameter. In order to work at a higher frequency and still produce large acoustic power, the transducer is excited with Q enhancement from resonance at the n th harmonic frequencies, $f_{n\text{th}}=v/\lambda_{n\text{th}}=nv/\lambda_{\text{fundamental}}=nf_{\text{fundamental}}$,

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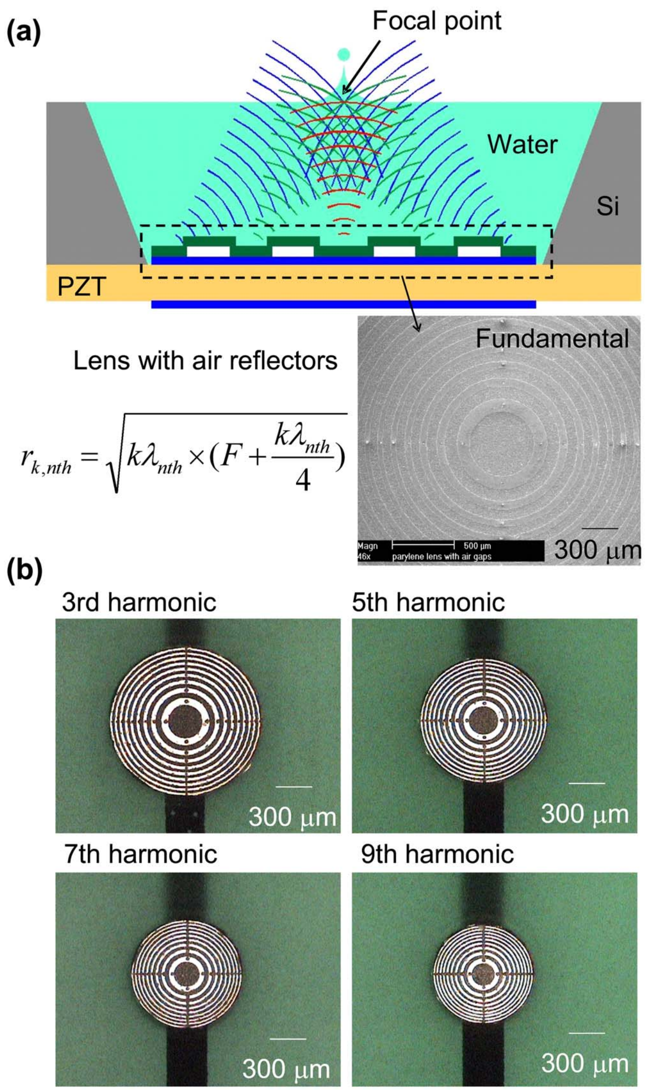

FIG. 1. (Color online) (a) Schematic diagram of the PZT acoustic ejector and scanning electron microscopy image of the acoustic lens. (b) Optical micrographs of the acoustic lenses for harmonic operations.

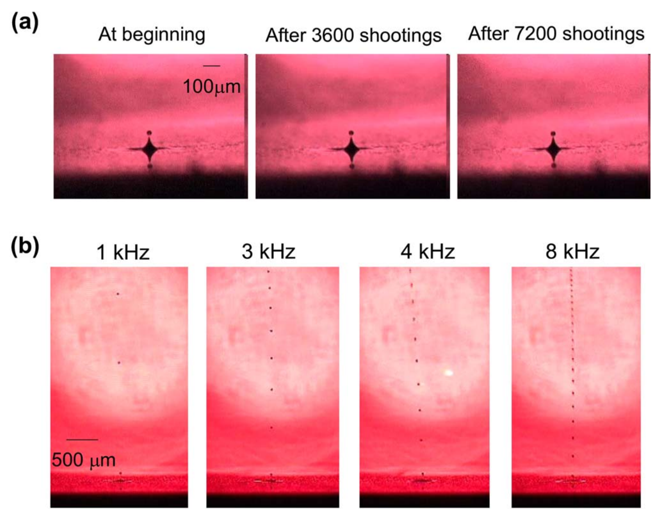

FIG. 2. (Color online) Droplet ejections by the ejector working at the third harmonic. (a) Optical micrographs of stable and continuous ejections. (b) Optical micrographs of ejections at high ejection rates.

where v is the acoustic velocity. For a symmetric transducer having electrodes sandwiching a piezoelectric sheet, there exist only odd harmonics, i.e., $n=1,3,5,\ldots$. With the fundamental frequency of 20 MHz, the third, fifth, seventh, and ninth harmonics are 60, 100, 140, and 180 MHz, respectively. Thus, through harmonic operations, a droplet of 10 μm in diameter can be theoretically obtained at the ninth harmonic frequency even with a 127 μm thick bulk PZT substrate.

The acoustic ejectors with the lenses designed for different harmonic operations have been fabricated. All ejectors were built on the same 127 μm thick PSI-5A4E PZT sheet (Piezo Systems, Cambridge, MA) but had different lens patterns. The PZT ejectors were adhesively bonded to microfluidic components (embedded microchannels, ejection chambers, and reservoirs), which were microfabricated with two silicon wafers. Figure 1(b) shows the optical micrographs of the fabricated acoustic lenses designed for different harmonics. For the acoustic ejectors containing the same number (nine) of Fresnel half-wave bands, the device footprint of the ejector working at the ninth harmonic is around ten times smaller than that of the ejector working at the fundamental resonant frequency. Thus, through harmonic operations, the acoustic transducer size can be advantageously reduced.

The fabricated ejector was driven with pulses of sinusoidal signals. Figure 2(a) shows the continuous ejection with ± 70 V_{pp} pulses of 58 MHz sinusoidal signals based on the third harmonic operation. The pulse width and the ejection rate are 8 μs and 120 Hz, respectively. The ejection is one droplet per pulse and free of satellite droplets. Frames at different times are almost identical, exhibiting the uniformity of the droplet sizes (26 μm in diameter) and the stability of the ejection process. Figure 2(b) shows the ejections at higher ejection rates. The images captured by strobing are actually a superposition of many successive droplets, and the image sharpness demonstrates consistent droplet ejections at the ejection rate up to 8 kHz.

The harmonic utilization for acoustic transducers has been observed to be applicable for ejection up to the ninth harmonic and eject uniform droplets down to 10 μm in diameter. Ejections for different harmonics have been thoroughly characterized and compared. Figure 3(a) shows the eminent droplet size reduction through harmonic operations. The measured droplet size is plotted as a function of the harmonic frequencies in Fig. 3(b). The theoretical wavelength in water is also plotted to demonstrate the droplet size's direct dependence on the wavelength. It is also noted that smaller droplets can be ejected within a shorter time. The measured droplet separation time t_{sep}, defined as the time it requires for the droplet to be formed and break free from the bulk liquid after the actuation signal is applied, is shorter for a smaller droplet size (Fig. 4). This is in good agreement with the theoretical and experimental results reported in Ref. 19, and thus higher ejection rates are possible with smaller droplets.

Compared with the nozzle-based ejectors, one of the challenges associated with the nozzleless ejector is the requirement of high-frequency electronics. For acoustic ejection, the droplet size is directly related to the acoustic wavelength, which is inversely proportional to the operation frequency. In order to eject the picoliter droplets, high-frequency electronics, including frequency synthesizers and amplifiers, are needed. The high-frequency electronic circuits are more complicated and more expensive than those

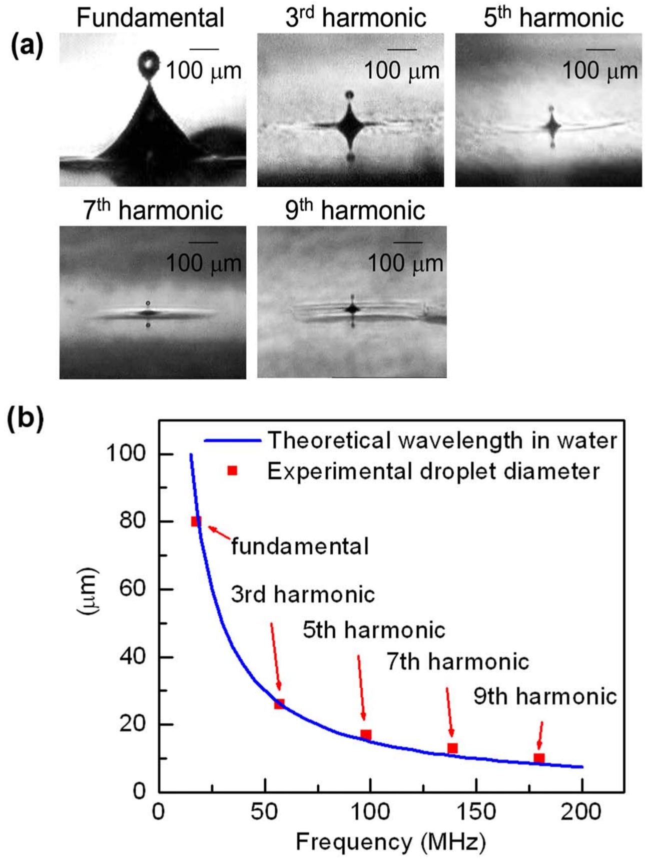

FIG. 3. (Color online) Droplet size reduction by harmonic operations. (a) Optical micrographs showing eminent droplet size reduction. (b) Experimental and theoretical curves of droplet size as a function of frequency.

typically used for nozzle-based inkjet printers. However, owing to the rapid progress of semiconductor technology, the high-frequency microelectronic devices are becoming more and more cost effective. Another point worth pointing out for the nozzleless ejection is the liquid evaporation. Since no nozzle is used for acoustic ejection and the droplets are ejected from open space, a certain mechanism would be needed to minimize the liquid evaporation. For example, a shutter can be utilized to dynamically open the ejection chamber during droplet ejection and to close the chamber in standby mode.

In conclusion, the use of harmonic frequencies to reduce the wavelength has been demonstrated for acoustic droplet ejection of subpicoliter liquid. The bulk acoustic transducer based on this idea is free of residual stress, robust, and reliable, yet efficient in producing large acoustic power to eject droplets down to 10 μm in diameter at the ninth harmonic (180 MHz). Consistent droplet ejections for different harmonic operations have been achieved at an ejection rate up to 8 kHz. The harmonic operations are excellent in reducing

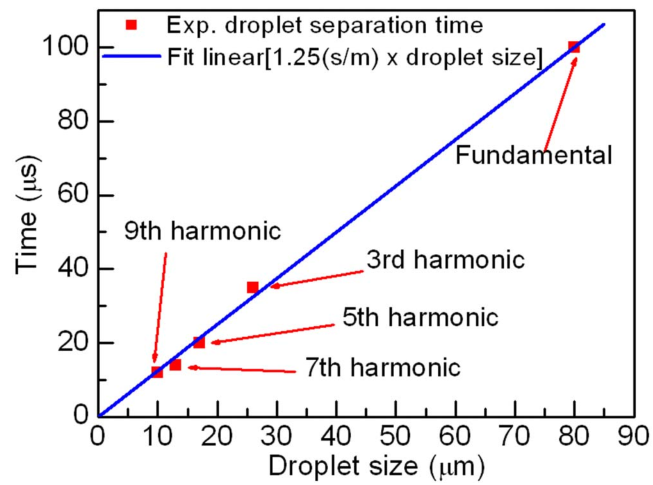

FIG. 4. (Color online) Measured droplet separation time vs droplet size.

not only the droplet size but also the transducer size. The latter has significant implication in microfluidic systems employing an array of acoustic transducers where the size of an individual transducer matters.

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